

## Spectral origins of giant Faraday rotation and ellipticity in Bi-substituted magnetic garnets

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Electrical permittivity theory has been applied to the interpretation of data describing the anomalously high Faraday rotation and ellipticity in the  $Y_{3-x}Bi_xFe_5O_{12}$  ferrimagnetic system. By the use of exact forms of the basic phenomenological equations for the off-diagonal tensor element  $\epsilon_1 = \epsilon'_1 + i\epsilon''_1$ , the measured  $Bi^{3+}$  influence on the separate component  $\epsilon'_1$  and  $\epsilon''_1$  spectra from 1 to 5 eV has been closely fitted to theory by the superposition of three  $Fe^{3+}$  (diamagnetic) electric dipole transitions occurring at 2.6, 3.15, and 3.9 eV, respectively. The strong transition at 3.15 eV that is believed to originate from  $Fe-O_6$  molecular complexes of the octahedral sublattice may also have a smaller companion peak in the vicinity of 5 eV. Transition bandwidths and excited-state splittings determined from the matching of theory to experiment both indicate that strong Bi covalent interactions exist with the  $Fe-O_6$  and  $Fe-O_4$  complexes. These results also confirm that the enhanced magneto-optical effects in the longer-wavelength region ( $\lambda \sim 1 \mu m$ ) are dominated by the tail of the  $\epsilon'_1$  diamagnetic peak at 3.15 eV.

## I. INTRODUCTION

The causes of magneto-optical effects on radiation in the infrared to ultraviolet bands have been a subject of increasing interest since the early works of Stephens,<sup>1</sup> Shen,<sup>2</sup> and Kahn *et al.*<sup>3</sup> More recently, the giant Faraday rotation in Bi-substituted magnetic garnets has breathed new life into the epitaxial garnet film technology. In spite of the efforts of many researchers, however, the origin of this anomaly remains uncertain. A major impediment to gaining insight into this phenomenon has been the lack of a physically realistic phenomenological interpretation of the reported spectra. This article describes the results of analyses based on the overlapping of three diamagnetic-type  $Fe^{3+}$  transitions that are influenced by covalent interactions with  $Bi^{3+}$  ions.

## II. THEORY

Magneto-optical interactions fall into two general categories: (i) paramagnetic, with a Zeeman-split ground state, and (ii) diamagnetic, with an excited state split  $2\Delta$  by spin-orbit coupling, as sketched in Fig. 1. For ferrimagnetic systems, Zeeman splitting is precluded by the strong superexchange field quenching of the spin degeneracy, leaving a spin singlet ground state. For this reason, interpretation of the rotation and ellipticity spectra from  $Y_{3-x}Bi_xFe_5O_{12}$  must logically be based on the diamagnetic functions described by the off-diagonal permittivity tensor elements  $\epsilon_1 = \epsilon'_1 + i\epsilon''_1$ , according to Allen and Dionne<sup>4</sup>

$$\epsilon_1 = \omega_p^2 \sum_{\pm} (\pm) \frac{f_{\pm}}{2\omega_{0\pm}} \times \frac{\omega(\omega_{0\pm}^2 - \omega^2 - \Gamma^2) + i\Gamma(\omega_{0\pm}^2 + \omega^2 + \Gamma^2)}{(\omega_{0\pm}^2 - \omega^2 + \Gamma^2)^2 + 4\omega^2\Gamma^2}, \quad (1)$$

where  $(\pm)$  indicates a subtraction,  $\omega_{0\pm} = \omega_0 \pm \Delta$ ,  $\omega_p^2 = 4\pi Ne^2/m$ , and  $N$  is the density of transition centers.

The oscillator strengths for the positive and negative rotations are  $f_+ \approx f_- \approx f/2$ , with  $f = (m\omega_0/h)(\langle g|x|e \rangle)^2$ , and  $x$  is the electric dipole operator. If we introduce the excited state splitting,  $f_{\pm} \approx (f/2)(1 \pm \Delta/\omega_0)$ .<sup>4</sup>

If this expression is separated into real and imaginary parts without approximations, one obtains separate relations for the  $\epsilon'_1$  and  $\epsilon''_1$  which may be used to compute Faraday and Kerr rotations and ellipticities.<sup>4</sup> For the magnetic garnets at energies below 2.5 eV, the diagonal elements of the tensor  $\epsilon'_0 \approx n^2$  ( $n$  is the index of refraction  $\approx 2.3$  in this regime) and  $\epsilon''_0 \approx 0$ . The Faraday rotation Verdet constant is given by

$$\theta_F \approx (\omega/2cn)\epsilon'_1. \quad (2)$$

For this system, both  $\epsilon'_0$  and  $\epsilon''_0$  are only modestly increased by  $Bi^{3+}$  substitutions. These features make Eq. (2) accurate for use in this energy regime.<sup>5</sup>

It is important to point out that the  $\epsilon'_1$  diamagnetic line shape features negative tails that result from the twin reverse peaks formed by the subtraction of the split dispersion curves offset by the energy  $2\Delta$ .<sup>4</sup>

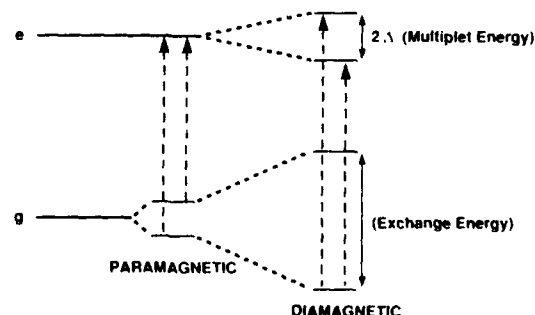
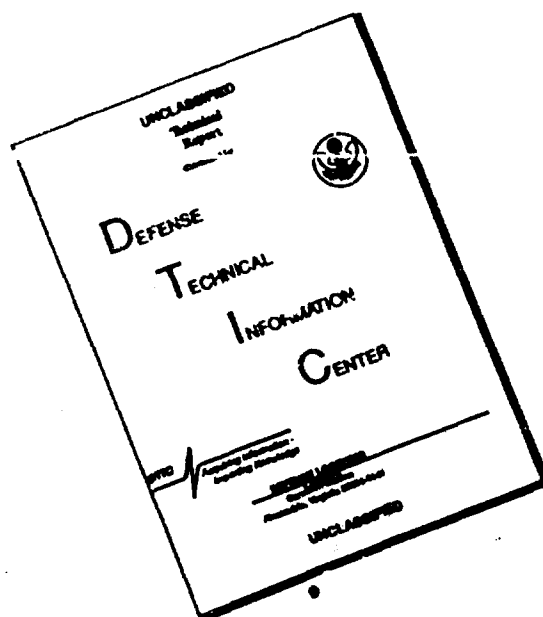


FIG. 1. Spectral transitions for the paramagnetic case, and the diamagnetic situation that arises in ferrimagnetism.

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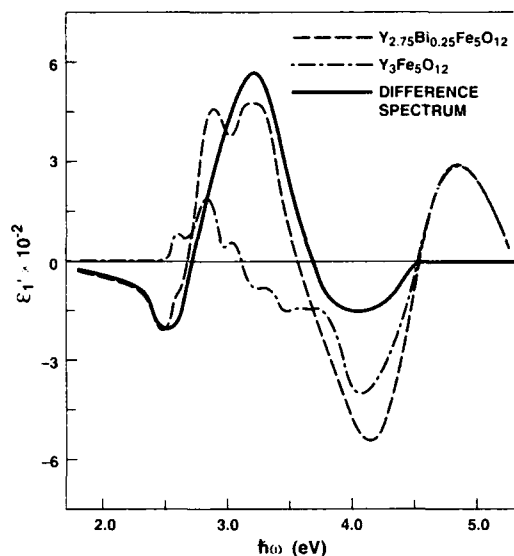


FIG. 2.  $Y_{3-x}Bi_xFe_5O_{12}$   $\epsilon_1''$  data of Wittekoek *et al.* (Ref. 5), showing the difference curve formed from the subtraction of the  $x=0$  and 0.25 curves.

### III. INTERPRETATION OF DATA

From inspection of the magnetic garnet magneto-optical data in the literature, one can identify two general groupings: (i) Kerr effect ellipsometry measurements of  $\epsilon_1'$  and  $\epsilon_1''$  as a function of spectral energy over the range from 2 to 5 eV, and (ii)  $\theta_F$  transmission measurements in the range below 2 eV. Although Faraday rotation effects are of practical importance at lower energies, the major optical events take place at higher energies. It is Kerr effect data, therefore, that provide the fundamental clues to the source of the phenomena. According to published data,<sup>5,6</sup> the major peak in  $\epsilon_1'$  lies between 3 and 3.5 eV.

In Fig. 2,  $\epsilon_1'$  data of  $Y_{3-x}Bi_xFe_5O_{12}$  that have been

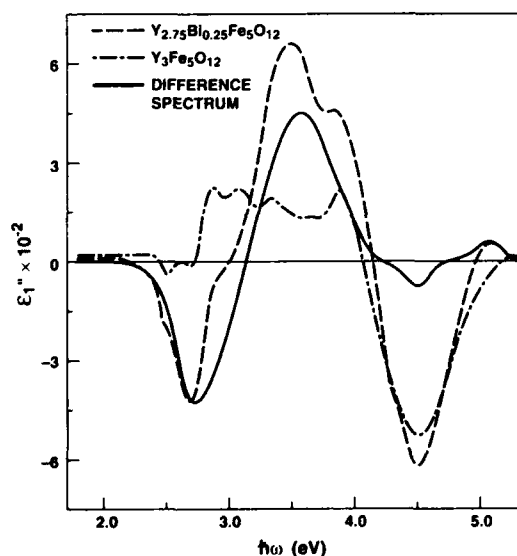


FIG. 3.  $Y_{3-x}Bi_xFe_5O_{12}$   $\epsilon_1''$  data of Wittekoek *et al.* (Ref. 5), showing the difference curve formed from the subtraction of the  $x=0$  and 0.25 curves.

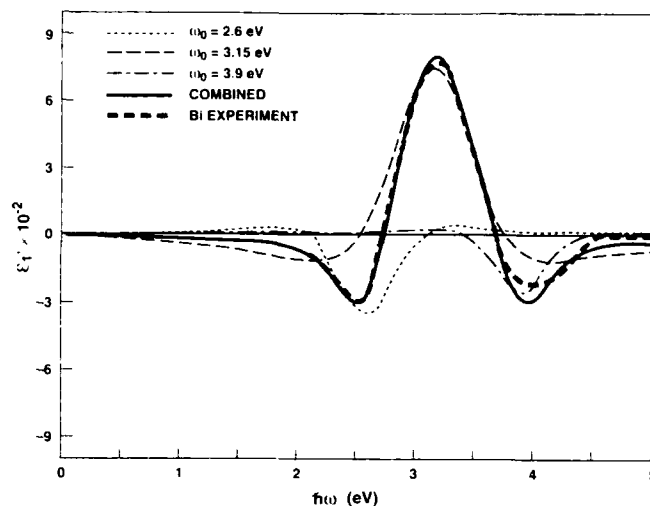


FIG. 4. Curves of  $\epsilon_1'$  calculated from the parameter values listed in Table I, showing the combined curve of the Bi effect between energies of 0 and 5 eV. The difference curve from Fig. 2 is added for comparison.

reduced from Kerr ellipsometry measurements by Wittekoek *et al.* are reproduced for  $x=0$  and 0.25. If the  $x=0$  (YIG) curve is treated as a base line, the curve for the Bi contribution is found by subtraction of the two curves. Figure 3 presents the results of the same procedure applied to the corresponding  $\epsilon_1''$  data. In both cases, the resultant curves reveal smooth Lorentzian-type functions of  $\omega$  depicting behavior that is strongly suggestive of two or three individual transition bands below 4 eV. The theory of Eq. (1) can be applied directly to the difference curves of Figs. 2 and 3 by selecting appropriate values of parameters  $\Delta$ ,  $\Gamma$ , and  $f$  for two principal diamagnetic transitions of opposite sign (the sign is determined by the direction of the magnetic moment that couples to the electric vector through spin-orbit coupling). As presented in Figs. 4 and 5, close fits to the experimentally derived curves are made with transitions at 2.6 and 3.15 eV over most of the range of

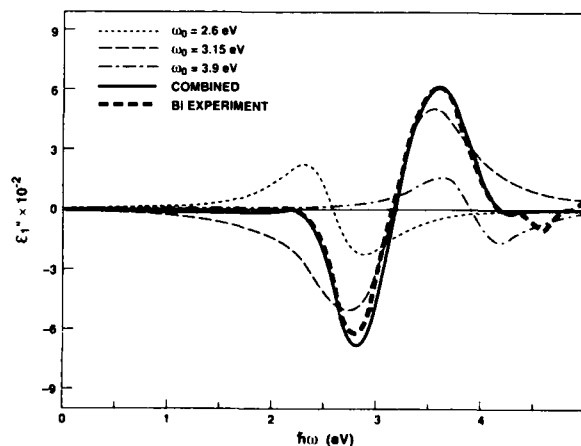


FIG. 5. Curves of  $\epsilon_1''$  calculated from the parameter values listed in Table I, showing the combined curve of the Bi effect between energies of 0 and 5 eV. The difference curve from Fig. 3 is added for comparison.

TABLE I. Spectral parameters of  $\epsilon'_1$  enhancement for  $x=0.25$ .

| Lattice site             | $\omega_0$<br>(eV) | $\omega_p^2 f$<br>(eV <sup>2</sup> ) | $\Gamma$<br>(eV) | $\Delta$<br>eV | $\Delta/\Gamma^b$ |
|--------------------------|--------------------|--------------------------------------|------------------|----------------|-------------------|
| Tetrahedral              | 2.6                | -2.8                                 | 0.44             | 0.11           | 0.25              |
| Octahedral               | 3.15               | 8                                    | 0.54             | 0.27           | 0.5               |
| Tetrahedral <sup>a</sup> | 3.9                | -3                                   | 0.44             | 0.11           | 0.25              |

<sup>a</sup>This transition appears to be of tetrahedral Fe origin but may also be influenced by charge-transfer excitations (which may not be Lorentzian) or by the Bi transition at 4.5 eV (Ref. 11).

<sup>b</sup>New data on saturation effects may determine that these values are appropriate for higher Bi concentrations.

measurement. To refine the interpretation and extend the model beyond 4 eV, a third transition of similar sign and proportions to the one at 2.6 eV is added at 3.9 eV.

It is also important to recognize that the unique spectral shapes of both  $\epsilon'_1$  and  $\epsilon''_1$  are fitted with the same set of parameters listed in Table I.

To highlight further the closeness of the fit between theory and experiment in the lower-energy region, the calculated curve for  $\epsilon'_1$  from Fig. 3 was modified according to Eq. (2), scaled to  $x=0.44$  (with the assumption that the  $\theta_F$  dependence on Bi content remains linear at small values of  $x$ ),<sup>4</sup> and plotted in Fig. 6 together with the corresponding  $\theta_F$  measured curve of Simsa *et al.*,<sup>7</sup> after subtraction of the  $Y_3Fe_5O_{12}$  baseline. The close agreement over the range from 1 to 2.5 eV indicates that the principal Bi contributions are not of paramagnetic origin.

#### IV. DISCUSSION

The opportunity to separate opposing magnetic sublattice contributions by the signs of the different Faraday peaks is an important aspect of magneto-optical spectra.

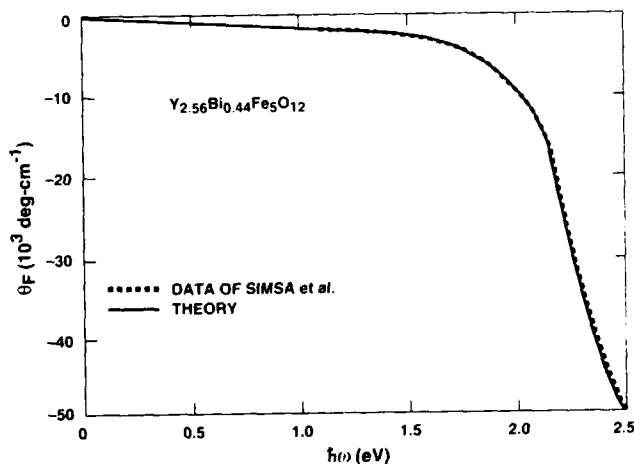


FIG. 6. Comparison between theory (scaled to  $x=0.44$ ) and experiment for energies below 2.5 eV, using parameter values of Table I. The data curve is from Ref. 7 and has also been corrected to remove the  $Y_3Fe_5O_{12}$  baseline.

Scott, Lacklison, and Page<sup>8</sup> concluded that the 3.15 eV line originates in the octahedral Fe-O<sub>6</sub> complexes (transitions from <sup>6</sup>S to excited <sup>4</sup>G or <sup>4</sup>D bands<sup>9</sup>). From Fig. 3, therefore, one concludes that the weaker 2.6 eV peak is of tetrahedral origin (Fe-O<sub>4</sub>). Most significant for practical matters is the dominance of the negative tail of the intense 3.15 eV line in the 1–2 eV region.

As listed in Table I, the  $\Gamma$  values for each transition are broad (0.25–0.5 eV). If the  $\epsilon''_0$  values are not greatly increased by Bi additions as reported,<sup>5</sup> the large enhancement of  $\epsilon'_1$  would not be caused by increased  $f$ . For the same reason, the proposition that Bi<sup>3+</sup> transitions cause the  $\epsilon_1$  anomalies would also be ruled out. For a homogeneous distribution, Bi<sup>3+</sup> ions could perturb and enhance the multiplet splitting of the excited band<sup>10</sup> of the exchange-coupled Fe<sup>3+</sup> lattices in direct proportion to the density and strength of the Fe<sup>3+</sup>-O<sup>2-</sup>-Bi<sup>3+</sup> bond linkages, thereby producing (to a first approximation) a linear growth in the product  $N\Delta$  with  $x$ , and a corresponding enhancement of  $\epsilon_1$  for selected Fe<sup>3+</sup> transitions. For fixed  $N$ , the largest reasonable  $\Delta/\Gamma$  values below saturation levels<sup>4</sup> were chosen to fit the data.

Reported saturation<sup>6</sup> of the  $\epsilon'_1$  peak at 3.15 eV as  $x \rightarrow 2$  may be explained by the ratio  $\Delta/\Gamma \rightarrow 1$ . This result further suggests that approximations based on the  $\Delta/\Gamma \ll 1$  assumption should be avoided in the interpretation of the Bi effects. It should be noted, however, that  $\epsilon'_1$  at fixed energies in the negative tails may not saturate because the smaller reverse peaks are moved to lower and higher energies, as illustrated in Allen and Dionne.<sup>4</sup> This latter feature would prove beneficial for applications that require materials with the highest  $\theta_F$  value at lower energies.

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